Spin and Charge Pumping by Ferromagnetic-Superconductor Order Parameters

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We study transport in ferromagnetic-superconductor/normal-metal systems. It is shown that charge and spin currents are pumped from ferromagnetic superconductors into adjacent normal metals by adiabatic changes in the order parameters induced by external electromagnetic fields. Spin and charge pumping identify the symmetry of the superconducting order parameter, e.g., singlet pairing or triplet pairing with opposite or equal spin pairing. Consequences for ferromagnetic-resonance experiments are discussed.

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Ferromagnetism induces a spin-dependent asymmetry in the densities of itinerant carriers. In contrast, superconductivity pairs electrons with equal or opposite spins depending on the symmetry of its order parameter. The coexistence of the two order parameters has been considered to be a rare phenomenon. However, recent experimental progress has demonstrated that ferromagnetism and superconductivity coexist in some materials like RuSr₂GdCu₂O₈ [1], UGe₂ [2], ZrZn₂ [3], and URhGe [4]. The experiments find triplet pairing in URhGe, and strong indications of triplet pairing in UGe₂ and ZrZn₂; they furthermore suggest that the same electrons are responsible for ferromagnetism as well as superconductivity. Besides, ferromagnetism and superconductivity are predicted to be simultaneously induced in hybrid ferromagnet (F)/normal-metal (N)/superconductor (S) systems [5]. These recent experimental discoveries, and the possibility of tailoring superconductivity and ferromagnetism in nanoscale systems, enable exploring novel physics involving pairing and spin-related transport processes. A variety of interesting spin phenomena have already been observed in hybrid F/N and semiconductor systems: e.g., giant-magnetoresistance—related effects, spin precession, and current-induced magnetization dynamics [6]. It is therefore natural to expect that interesting rich phenomena should also occur in FS/N systems.

This Letter demonstrates how the coexistence of superconductivity and ferromagnetism is manifested in the adiabatic pumping in hybrid FS/N structures. In particular, we study spin and charge pumping when the magnetization slowly precesses, which can be achieved in ferromagnetic resonance (FMR) and in current-induced magnetization dynamics. FMR experiments have already been carried out to investigate the magnetism in RuSr₂GdCu₂O₈ [7]. We also consider pumping by slow variations in the phase of the singlet or triplet order parameter, or in the direction of the triplet order parameter. These can be induced by electric and magnetic fields to be discussed below. By pumping we thus mean the spin and charge flows into the adjacent normal conductors in response to adiabatic changes in the FS order parameters.

Consequently, in the case of pumping by the magnetization direction, we compute the spin current \mathbf{I}_s and the charge current I_c for a given rate of the magnetization-direction change, and for pumping by changing pairing, we compute the same quantities as functions of the phase-change or the direction-change rates of the pair correlations. We employ two approaches giving identical results: 1) solving the time-dependent ac problem directly and 2) using a gauge transformation to obtain a time-independent dc problem. We first explain our model and notation, before proceeding to the derivation and results. Experimental consequences are discussed in the end.

A ferromagnetic superconductor is treated in the mean-field approximation, where ferromagnetism is represented by the average magnetization and superconductivity is described by a pair potential. Our model is phenomenological and we do not discuss the microscopic origin of the exchange field or the superconducting pairing. The Bogoliubov-de Gennes (BdG) equation is

$$\begin{pmatrix} \hat{\xi} & \hat{\Delta} \\ -\hat{\Delta}^* & -\hat{\xi}^* \end{pmatrix} \begin{pmatrix} \hat{u} \\ \hat{v} \end{pmatrix} = i\hbar \frac{\partial}{\partial t} \begin{pmatrix} \hat{u} \\ \hat{v} \end{pmatrix}, \tag{1}$$

where $\hat{\xi} = H_0 \hat{1} + \hat{\boldsymbol{\sigma}} \cdot \boldsymbol{\epsilon}_{xc}$ is the single-particle Hamiltonian, $\epsilon_{xc} = \epsilon_{xc} \mathbf{m}$ (with $\epsilon_{xc} > 0$) is the ferromagnetic exchange field along the magnetization direction \mathbf{m} and $\hat{\Delta} = (d_0 \hat{1} + \hat{\boldsymbol{\sigma}} \cdot \boldsymbol{d}) i \hat{\sigma}_y$ is the superconducting pair potential, in terms of the singlet (scalar) part, d_0 , and the triplet (vector) part, $\mathbf{d} = (d_x, d_y, d_z)$. $\hat{u}^T = (u^{\uparrow}, u^{\downarrow})$ are spin-dependent electron wave functions and $\hat{v}^T = (v^{\uparrow}, v^{\downarrow})$ are those of holes; $\hat{\boldsymbol{\sigma}} = (\hat{\sigma}_x, \hat{\sigma}_y, \hat{\sigma}_z)$ is the vector of Pauli matrices. The single-particle Hamiltonian H_0 contains the kinetic and potential-energy terms. The Fermi energy is taken to be the largest relevant energy scale. The local exchange field can be position dependent close to the interface, $\epsilon_{xc}(\mathbf{r})$, and the pair potential $\hat{\Delta}(\mathbf{k},\mathbf{r})$ can also be position as well as wave-vector, **k**, dependent [8]. For simplicity, we assume that the exchange field and the pair potential are uniform inside the superconductor and drop to zero at the FS/N interface: $\epsilon_{xc}(\mathbf{r}) = \epsilon_{xc}\Theta(z)$ and $\hat{\Delta}(\mathbf{k},\mathbf{r}) = \hat{\Delta}(\mathbf{k})\Theta(z)$, where $\Theta(z)$ is the Heaviside

step function and z is the coordinate perpendicular to the FS/N interface.

Fermionic statistics dictates $\hat{\Delta}(\mathbf{k}) = -\hat{\Delta}^T(-\mathbf{k})$ [9]. The singlet (triplet) part of $\hat{\Delta}$ thus needs to have even (odd) parity: $d_0(\mathbf{k}) = d_0(-\mathbf{k})$ and $\mathbf{d}(\mathbf{k}) = -\mathbf{d}(-\mathbf{k})$. We study in the following two simple cases of triplet superconductors: opposite-spin pairing (OSP) along the exchange field, $\mathbf{d}(\mathbf{k}) \times \epsilon_{xc} = 0$ and equal-spin pairing (ESP), $\mathbf{d}(\mathbf{k}) \cdot \boldsymbol{\epsilon}_{xc} = 0$ [10]. Triplet OSP superconductors are described by a (complex-valued) vector $\mathbf{d}(\mathbf{k}) = d(\mathbf{k})\mathbf{m}$ along the magnetization direction. We show that the transport properties in triplet OSP are similar to those of singlet pairing. In ESP, superconductivity occurs independently for spins along and opposite to the magnetization direction: By choosing the magnetization along the zaxis, the superconducting pair potential decomposes into two terms, corresponding to spins up and down along the z axis, $d^{\uparrow(\downarrow)}(\mathbf{k}) = \mp d_x(\mathbf{k}) + i d_y(\mathbf{k})$. Since superconducting correlations do not mix the spin-up and down subsystems, two ESP phases can be distinguished: the A_1^{σ} phase, where pairing occurs only for spin σ (i.e., $d^{\sigma} \neq 0$ and $d^{-\sigma} = 0$) and the A₂ phase, where pairing occurs for both spins. A large exchange interaction, $\epsilon_{\rm xc} \gtrsim |d_0|$ (|d|), suppresses superconducting singlet (triplet) OSP correlations [10]. We therefore only consider $\epsilon_{xc} < |d_0|$ (|d|) for these systems, so that the quasiparticle excitations have a finite gap. Triplet ESP have a quasiparticle gap in the superconducting spin channels independent of the size of the exchange interaction [10], and we do not make assumptions about the ratio of the exchange field to the pair potential in this case.

Let us apply the standard scattering-matrix approach [11, 12, 13, 14, 15] to an FS/N system. We assume that the Hamiltonian H_0 is continuous across the FS/N interface and incorporate interfacial disorder and bandstructure mismatch into a "disordered" normal region [11]. Similarly to Ref. [11], we solve the BdG equation for an electron (or hole) incident on the specular FS/N interface from the normal-metal side. The total reflection matrix is then found by concatenating the FS/N reflection with the scattering by the normal disordered region for electrons and holes. The FS layer, in series with the disordered region, is viewed as a scatterer for electrons supplied by the normal reservoir, see Fig. 1. The problem is simplified in the clean-superconductor limit, where the FS mean free path is longer than the superconducting coherence length $\hbar v_F/(\pi \Delta)$, expressed in terms of the Fermi velocity v_F and quasiparticle gap Δ . Focusing on the low-temperature regime, $k_BT \ll \Delta$, we define the $M \times M$ spin-dependent electron \rightarrow electron and electron—hole reflection matrices r_{ee}^{σ} and r_{he}^{σ} , where Mis the total number of quantum channels, or transverse waveguide modes, at the Fermi level of the normal-metal lead and σ is the spin label along the magnetization direction for incident electrons. For singlet or triplet OSP, r_{he}^{σ} describes reflection into holes with spin $-\sigma$, while

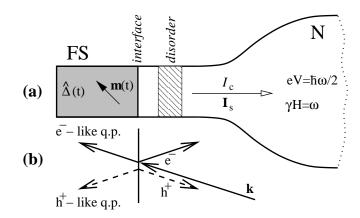


FIG. 1: (a) A ferromagnetic superconductor coupled to a normal reservoir through a specular FS/N interface in series with a normal disordered region. (b) Electrons incident on the FS/N interface from the normal side are reflected as electrons or holes and transmitted as decaying electron- or hole-like quasiparticles.

for triplet ESP, reflected holes have the same spin σ as incident electrons.

We find that for both dc and adiabatic ac transport, spin and charge currents are uniquely determined by two real-valued conductances and one complex-valued conductance, representing transport of spins aligned to the magnetization, and transverse to the magnetization, respectively:

$$g^{\uparrow\uparrow} = \operatorname{Tr} \left[r_{he}^{\uparrow} (r_{he}^{\uparrow})^{\dagger} \right], \ g^{\downarrow\downarrow} = \operatorname{Tr} \left[r_{he}^{\downarrow} (r_{he}^{\downarrow})^{\dagger} \right], \quad (2)$$

$$g^{\uparrow\downarrow} = \text{Tr} \left[1 - r_{ee}^{\uparrow} (r_{ee}^{\downarrow})^{\dagger} + (r_{he}^{\uparrow})^{\dagger} r_{he}^{\downarrow} \right].$$
 (3)

It is convenient to define the total conductance $g = g^{\uparrow\uparrow} + g^{\downarrow\downarrow}$ and the polarization $p = (g^{\uparrow\uparrow} - g^{\downarrow\downarrow})/g$. We will below interpret how these conductances determine charge and spin flow, and discuss their values in various limits.

Let us first fix the magnetization direction \mathbf{m} and consider pumping by an adiabatically varying phase $\phi(t)$ of the superconducting pair potential, $\partial_t \phi = \omega \ll (\epsilon_{\rm xc}, \Delta)/\hbar$. As is well known, this time-dependent problem can be transformed into a dc problem by a gauge transformation $U(t) = e^{i\phi(t)/2}$: Varying the phase results in nonequilibrium transport corresponding to a dc response at a voltage bias $\hbar\omega/(2e)$. The pumped current is thus nothing more than a response to a voltage $V = \hbar\omega/(2e)$ applied between the superconducting condensate and the normal metal, and we compute

$$I_c = \frac{e}{2\pi} g\omega \text{ and } \mathbf{I}_s = -\frac{\hbar}{4\pi} pg\omega \mathbf{m}.$$
 (4)

g is thus the usual Andreev-reflection conductance, in units of $e^2/(\pi\hbar)$. The spin current is determined by the conductance polarization p. Eqs. (4) can also be derived in the ac pumping framework of Ref. [15].

Pure spin flow is generated by the variations in the magnetization direction **m**, induced by, e.g., a resonant rf magnetic field. In Ref. [14] we computed the corresponding pumped spin flow into the normal metal,

$$\mathbf{I}_s = \frac{\hbar}{4\pi} \left(\operatorname{Re} g^{\uparrow\downarrow} \mathbf{m} \times \frac{d\mathbf{m}}{dt} + \operatorname{Im} g^{\uparrow\downarrow} \frac{d\mathbf{m}}{dt} \right) , \qquad (5)$$

for a general F/N contact with no superconducting correlations. Here $g^{\uparrow\downarrow} = \text{Tr}[1 - r_{ee}^{\uparrow}(r_{ee}^{\downarrow})^{\dagger}]$ is the F/N mixing conductance for transverse spins expressed in terms of the normal-side reflection matrices [13]. The pumped spin flow induces an enhanced Gilbert damping when the normal metal is a good spin sink, so that $g^{\uparrow\downarrow}$ is experimentally measurable [14]. We have generalized Eq. (5) to spin pumping by a superconducting ferromagnet: The pumped spin flow remains of the form (5) with a redefined mixing conductance $g^{\uparrow\downarrow}$ (3). There is no accompanying charge pumping. We derive Eqs. (3) and (5) by two different methods: First, we extend the pumping approach of Ref. [14], where the scattering matrix is time-dependent in spin space due to a slow variation in the magnetization direction m, to include electron→hole reflection. Secondly, and much simpler, the calculation is reformulated as a dc problem in the spin frame of reference which is moving together with $\mathbf{m}(t)$. The latter is achieved by (instantaneously) applying a spin-rotation operator around the vector $\mathbf{m} \times \partial_t \mathbf{m}$ and correspondingly adding a new term in the Hamiltonian: $\hat{\xi}' = -(\hbar/2)(\mathbf{m} \times \partial_t \mathbf{m}) \cdot \hat{\boldsymbol{\sigma}}$. This term corresponds to an equilibrium transverse spin accumulation which, in turn, induces the spin current (5). The mixing conductance (3) is obtained after extending the F/N dc theory of Ref. [13] to account for electron—hole reflection. By unitarity of the scattering matrix [11], the real part of $g^{\uparrow\downarrow}$ is bounded from above by twice the number of channels: $\operatorname{Re} g^{\uparrow\downarrow} \leq 2M$.

For triplet OSP superconductors, the derivation leading to Eq. (5) assumes that the triplet pair-potential direction moves together with m, while its wave-vector dependence is locked to the atomic lattice: $d(\mathbf{k},t) =$ $d(\mathbf{k})\mathbf{m}(t)$. For triplet ESP, the situation is more complex since the vector pair potential d is perpendicular to \mathbf{m} , resulting in an additional dynamic degree of freedom. In particular, the magnetization motion does not uniquely determine the trajectory of d. In deriving Eq. (5), we assumed that d rotates together with m around m's instantaneous rotation axis. We can get more complex trajectories by combining (instantaneous) rotations of m and d with "twisting" of d around m and the overall phase variation of the pair potential. The induced currents will then be given by adding the corresponding contributions to pumping. Calculating the exact trajectories for realistic systems, which might be governed by spin-orbit interactions in the lattice field or other microscopic details, lies beyond the scope of this paper. A simple example is the clockwise rotation of d around m with frequency ω

which induces additional currents

$$I_c = -\frac{e}{2\pi}pg\omega \text{ and } \mathbf{I}_s = \frac{\hbar}{4\pi}g\omega\mathbf{m}.$$
 (6)

Note that while the preceding equations are general, Eq. (6) applies to the case of triplet ESP only. For a FS/N system disconnected from an Ohmic circuitry, the low-frequency charge flow vanishes, so that the overall phase variation of \boldsymbol{d} and its twisting around \boldsymbol{m} must result in canceling charge currents, I_c 's, in Eqs. (4) and (6), but a finite net spin current for p < 1. Eqs. (6) can be derived similarly to Eq. (5), either as a time-dependent pumping problem or a dc problem in the gauge-transformed frame of reference rotating with \boldsymbol{d} .

Eqs. (4), (5), and (6) are general expressions for pumped charge and spin flows by varying FS order parameters. These currents are all governed by two real-valued and one complex conductance parameters: $q^{\uparrow\uparrow}$, $q^{\downarrow\downarrow}$, and $q^{\uparrow\downarrow}$, which can be evaluated in microscopic models. A finite value of the electron—hole reflection coefficient, r_{he}^{σ} , requires that an electron incident from the normal-metal reservoir gets transmitted through the disordered region, converted into holes at the interface, and transmitted back into the normal lead as a hole, see Fig. 1. For small (characteristic) transmission eigenvalues T of the normal disordered region, $g^{\uparrow\uparrow}$ and $g^{\downarrow\downarrow}$ therefore scale as T^2 rather than as T, as in the Landauer-Büttiker formula for an N/N contact. Indeed, the Andreev conductance for a singlet nonmagnetic-superconductor/normal-metal contact was shown to be $g_{\rm S/N} = \sum_m 2T_m^2/(2-T_m)^2$, where m labels the transmission eigenvalues for scattering in the normal metal [11]. In the limit of no disorder (and no band-structure mismatch), $T_m \equiv 1$ and $g_{S/N} = 2M$, i.e., Andreev reflection causes a doubling of the conductance, as compared to the N/N interface. We generalize $q_{S/N}$ to the case of a magnetic singlet superconductor with an s-wave symmetry of the pair potential, $d_0(\mathbf{k}) \equiv |d_0|e^{i\phi}$:

$$g = \sum_{m} \frac{2T_m^2}{(2 - T_m)^2 - 4(1 - T_m)(\epsilon_{xc}/|d_0|)^2}$$
 (7)

and p=0. The mixing conductance (3) of s-wave magnetic superconductors can also be expressed in terms of the scattering matrix of the disordered region, by generalizing the formalism of Ref. [11]. For that we need to concatenate the transfer matrix of the normal disordered region with electron hole conversion at the interface: $r_{eh(he)}^{\uparrow} = e^{i(\beta \pm \phi)}$ and $r_{eh(he)}^{\downarrow} = e^{-i(\beta \mp \phi)}$, where $\beta = \arccos(\epsilon_{\rm xc}/|d_0|)$. The situation is even more complicated for the k-dependent pair potential (which is always the case for the odd-parity triplet pairing) and we do not pursue it here. The exception is the case with no disordered region in our model, Fig. 1. The conductance parameters for the singlet and triplet OSP in such bal-

listic systems are then given by

$$g = \frac{Ak_F^2}{2\pi}, \ p = 0, \text{ and}$$
 (8)

$$g^{\uparrow\downarrow} = A \int \frac{d^2 \mathbf{k}_{\perp}}{(2\pi)^2} \left\{ 1 \pm \exp\left[-2i\arccos\frac{\epsilon_{\text{xc}}}{\Delta(\mathbf{k})}\right] \right\}, (9)$$

where the \pm sign corresponds to singlet (triplet) pairing and $\Delta = |d_0|$ or |d|, respectively. \mathbf{k} is the incident-electron wave vector at the Fermi level, $|\mathbf{k}| = k_F$, and \mathbf{k}_{\perp} is its transversal projection in the lead with cross section A. $r_{he}^{\uparrow} = e^{i(\beta-\phi)}$ and $r_{he}^{\downarrow} = -e^{-i(\beta+\phi)}$, where $\beta = \arccos(\epsilon_{\rm xc}/|d|)$ for the triplet OSP with $d = |d|e^{i\phi}$, for a given \mathbf{k} .

It is instructive to discuss the mixing conductance $q^{\uparrow\downarrow}$ (3) values in some special cases of a clean interface with matched band structures since it can be measured experimentally in FMR. For an F/N interface, $r_{ee}^{\sigma}=r_{he}^{\sigma}=0$, while $g^{\uparrow\downarrow}=M$ is large determined by the number of transverse wave-guide channels M, assuming that the F layer is thicker than the ferromagnetic coherence length $\hbar v_F/(\pi \epsilon_{\rm xc})$. For a perfect electron—hole reflection off the singlet superconductor, we find $r_{he}^{\uparrow} = -r_{he}^{\downarrow}$, $|r_{he}^{\sigma}| = 1$, in the limit when $\epsilon_{xc} \ll \Delta$, resulting in a vanishing mixing conductance, as follows from Eq. (9). This is easy to understand since $r_{he}^{\uparrow} = -r_{he}^{\downarrow}$ means that the transverse spin-↑ electrons get reflected as the spin-↓ holes which exactly cancel the incident transverse spin current. In the analogous limit of the triplet OSP, $r_{he}^{\uparrow} = r_{he}^{\downarrow}$, $|r_{he}^{\sigma}| = 1$, doubling the F/N mixing conductance. We thus find that $g^{\uparrow\downarrow} = M$, 0, 2M for F/N, singlet FS/N, and triplet OSP FS/N interfaces. Since $g^{\uparrow\downarrow}$ is a direct measure of the ferromagnetic Gilbert-damping enhancement [14], the onset and the nature of the superconducting pairing has nontrivial consequences in FMR experiments [7]. It is worthwhile noting that $g^{\uparrow\downarrow} \to 2M$ is a rather special limit for the mixing conductance in multilayer magnetoelectronic circuit theories [16, 17]. For example, in the case of a symmetric FS/N/FS structure, it should result in a dynamic locking of the two magnetizations which can be measured experimentally [17].

Finally, in triplet ESP with no disordered region,

$$g = \frac{A k_F^2}{4\pi} \,, \ p = \sigma \,, \ \text{and} \ g^{\uparrow\downarrow} = g \eqno(10)$$

in the A_1^{σ} phase, and

$$g = \frac{Ak_F^2}{2\pi}, \ p = 0, \text{ and}$$
 (11)

$$g^{\uparrow\downarrow} = A \int \frac{d^2 \mathbf{k}_{\perp}}{(2\pi)^2} \left\{ 1 + \exp\left(i[\phi^{\uparrow}(\mathbf{k}) - \phi^{\downarrow}(\mathbf{k})]\right) \right\} (12)$$

in the A_2 phase. $r_{he}^{\sigma} = ie^{-i\phi^{\sigma}}$, for each superconducting spin channel with $d^{\sigma} = |d^{\sigma}|e^{i\phi^{\sigma}} \neq 0$. Note that in deriving Eq. (5), we made a convention for the instantaneous coordinate system of $\hat{\mathbf{x}} \propto \mathbf{m} \times \partial_t \mathbf{m}$, $\hat{\mathbf{y}} \propto -\partial_t \mathbf{m}$,

and $\hat{\mathbf{z}} = \mathbf{m}$. [It is necessary to specify the coordinate-system convention in the case of the A_2 phase, because $d^{\uparrow(\downarrow)} = \mp d_x + i d_y$ and, since \mathbf{d} transforms as a vector, the relative phase of $d^{\uparrow(\downarrow)}$ entering Eq. (12) depends on the choice of the x and y axes.] It then follows that the second term in the curly brackets of Eq. (12) is modulated for a small-angle precession of \mathbf{m} , unless \mathbf{d} twists around \mathbf{m} with its precession frequency (i.e., following the instantaneous rotation axis). In the former case, the theory therefore predicts an anisotropic Gilbert-damping parameter. For the triplet ESP A_2 phase, the symmetry of the superconducting order parameter may thus be seen in an additional anisotropic FMR line width, as one changes the temperature across the F-to-FS transition.

In summary, we have studied the interplay between ferromagnetism and superconductivity in adiabatic pumping by varying order parameters in FS materials in contact with normal metals. We demonstrate that the symmetry of the superconducting pair potential is reflected in the conductance parameters which govern the pumped spin and charge flows. An experimental quantity of a particular interest, which encodes information about both ferromagnetic and superconducting correlations, is the mixing conductance $g^{\uparrow\downarrow}$ which governs the Gilbert damping of the magnetization dynamics [14]. Consequently, signatures of the FS order parameter can be measured in thin film FS/N or FS/N/FS FMR experiments.

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